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ELAM-T LIMITED 103 BOROUGH ROAD LONDON SE1 OAA

Patents ADP number (if you know it)

If the applicant is a corporate body, give the country/state of its incorporation

08105280001

Title of the invention

ELECTROLUMINESCENT MATERIALS AND DEVICES

5. Name of your agent (if you bave one)

"Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)

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Number of earlier application

Date of filing
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Claim(s) 4

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A. N. Cohen

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Electroluminescent Materials and Devices

The present invention relates to electroluminescent materials and to electroluminescent devices.

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Materials which emit light when an electric current is passed through them are well known and used in a wide range of display applications. Liquid crystal devices and devices which are based on inorganic semiconductor systems are widely used, however these suffer from the disadvantages of high energy consumption, high cost of manufacture, low quantum efficiency and the inability to make flat panel displays.

Organic polymers have been proposed as useful in electroluminescent devices, but it is not possible to obtain pure colours, they are expensive to make and have a relatively low efficiency.

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Another compound which has been proposed is aluminium quinolate, but this requires dopants to be used to obtain a range of colours and has a relatively low efficiency.

Patent application WO98/58037 describes a range of lanthanide complexes which can be used in electroluminescent devices which have improved properties and give better results. Patent Applications PCT/GB98/01773, PCT/GB99/03619, PCT/GB99/04030, PCT/GB99/04024, PCT/GB99/04028, PCT/GB00/00268 describe electroluminescent

complexes, structures and devices using rare earth chelates.

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US Patent 5128587 discloses an electroluminescent device which consists of an organometallic complex of rare earth elements of the lanthanide series sandwiched between a transparent electrode of high work function and a second electrode of low work function with a hole conducting layer interposed between the

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electroluminescent layer and the transparent high work function electrode and an electron conducting layer interposed between the electroluminescent layer and the electron injecting low work function anode. The hole conducting layer and the electron conducting layer are required to improve the working and the efficiency of the device. The hole transporting layer serves to transport holes and to block the electrons, thus preventing electrons from moving into the electrode without recombining with holes. The recombination of carriers therefore mainly takes place in the emitter layer.

We have now invented electroluminescent compounds and devices incorporating them.

According to the invention there is provided electroluminescent compounds of formula $M(L)_n$ and $MO(L)_{n-2}$ where M is a metal in a valency state n of greater than 3 and L is an organic ligand, the ligands L can be the same or different e.g. $M(L_1)$ (L₂) (L₃) (L₄)... or $MO(L_1)$ (L₂)....

Preferably the metal M is a transition metal such as titanium, zirconium or hafnium in the four valency state or vanadium, niobium or tantulum in the five valency state.

Preferably the electroluminescent compound is doped with a dopant preferably in an amount of 5 to 15% of the doped mixture.

Dopants which can be used include diphenylacridine, coumarins, perylene and their derivatives.

The preferred dopants are coumarins such as those of formula

$$R_1$$
 R_2
 R_3
 R_2
 R_3
 R_4
 R_4
 R_5
 R_6

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where R_1 , R_2 , and R_3 are hydrogen or an alkyl group such as a methyl or ethyl group, amino and substituted amino groups e.g.

$$R_{1}$$
 R_{2}
 R_{3}
 R_{3}
 R_{3}
 R_{3}
 R_{3}

where R₃ is hydrogen or alkyl group such as a methyl or ethyl group,

Examples of coumarins are given in figs. 9 and 10 of the drawings

Other dopants include salts of bis benzene sulphonic acid such as

15 (C)

and perylene and perylene derivatives and dopants of the formulae of figs. 11 to 13 of the drawings where R₁, R₂, R₃ and R₄ are R, R₁, R₂, R₃ and R₄ can be the same or different and are selected from hydrogen, hydrocarbyl groups, substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorocarbons

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such as trifluoryl methyl groups, halogens such as fluorine or thiophenyl groups; R_1 , R_2 , R_3 and R_4 can also form substituted and unsubstituted fused aromatic, heterocyclic and polycyclic ring structures and can be copolymerisable with a monomer e.g. styrene. R_1 , R_2 , R_3 and R_4 can also be unsaturated alkylene groups such as vinyl groups or groups

$$--$$
C $--$ CH $_2$ $--$ R

where R is as above.

The formulae of examples of the complexes are shown in fig. 1a and fig. 1b of the accompanying drawings where M is titanium, zirconium or hafnium in the four valency state and (L₁), (L₂), (L₃) and (L₄) can be the same of different and can form fused cyclic, heterocyclic, aromatic or substituted aromatic rings or fig 1c and 1d where M is vanadium, niobium or tantulum in the five valency state and (L₁), (L₂), (L₃) (L₄) and (L₅) can be the same of different and can form fused cyclic, heterocyclic, aromatic or substituted aromatic rings.

Other preferred ligands L are quinolate, porphoryin, porphines, pyrazalones, such as 5,10,15,20-tetra(4-pyridyl) 21H,23H-porphine 5,10,15,20-tetra(p-tolyl) 21H,23H-porphine and meso- tetraphenyl porphine and β diketones such as dibenzoyl methane and ligands of formula

$$\begin{pmatrix}
R_1 \\
R_2 \\
R_3
\end{pmatrix}$$
or
$$\begin{pmatrix}
R_1 \\
Y \\
R_3
\end{pmatrix}$$
or
$$\begin{pmatrix}
R_1 \\
R_2 \\
R_3
\end{pmatrix}$$
(II)
$$\begin{pmatrix}
R_1 \\
R_2 \\
R_3
\end{pmatrix}$$
(III)

where R_1 , R_2 and R_3 can be the same or different and are selected from hydrogen, and substituted and unsubstituted hydrocarbyl groups such as substituted and unsubstituted aliphatic groups, substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorocarbons such as trifluoryl methyl groups, halogens such as fluorine or thiophenyl groups; R_1 , R_2 and R_3 can also form

substituted and unsubstituted fused aromatic, heterocyclic and polycyclic ring structures and can be copolymerisable with a monomer e.g. styrene. X is Se, S or O, Y can be hydrogen, substituted or unsubstituted hydrocarbyl groups, such as substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorine, fluorocarbons such as trifluoryl methyl groups, halogens such as fluorine or thiophenyl groups or nitrile.

Examples of R₁ and/or R₂ and/or R₃ include aliphatic, aromatic and heterocyclic alkoxy, aryloxy and carboxy groups, substituted and substituted phenyl, fluorophenyl, biphenyl, phenanthrene, anthracene, naphthyl and fluorene groups alkyl groups such as t-butyl, heterocyclic groups such as carbazole.

In dibenzoyl methane in (I) R_1 and R_3 are phenyl groups and R_2 is hydrogen. Other ligands L can be

um Complexes 2 11:39

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where the gropus R can be the same or different and are selected from hydrogen, and substituted and unsubstituted hydrocarbyl groups such as substituted and unsubstituted aliphatic groups, substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorocarbons such as trifluoryl methyl groups, halogens such as fluorine or thiophenyl groups; R₁, R₂ and R₃ can also form substituted and unsubstituted fused aromatic, heterocyclic and polycyclic ring structures and can be copolymerisable with a monomer e.g. styrene.

Some of the different groups L may also be the same or different charged groups such as carboxylate groups so that the group L_1 can be as defined above and the groups L_2 , L_3 ... can be charged groups such as

$$R - C$$

where R is R_1 as defined above or the groups L_1 , L_2 can be as defined above and $L_{3...}$ etc. are other charged groups.

R₁, R₂ and R₃ can also be

A preferred moiety R₁ is trifluoromethyl CF₃ and examples of such diketones are, banzoyltrifluoroacetone, p-chlorobenzoyltrifluoroacetone, p-bromotrifluoroacetone, p-phenyltrifluoroacetone, 1-naphthoyltrifluoroacetone, 2-naphthoyltrifluoroacetone, 2-phenathoyltrifluoroacetone, 3-phenanthoyltrifluoroacetone, 9-anthroyltrifluoroacetonetrifluoroacetone, cinnamoyltrifluoroacetone, and 2-thenoyltrifluoroacetone.

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The different groups L may be the same or different ligands of formulae

$$\begin{pmatrix} R_1 & X & X \\ R_3 & X & X \\ R_2 & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & & \\ & & & & \\ & & & & \\ & & & & \\ & & & & \\ & & & & \\ & & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & \\ & & & \\ & \\ & & \\ & & \\ & \\ & & \\ & \\ & & \\ & \\ & \\ & & \\ & \\ & \\ & & \\ & \\ & \\$$

where X is O, S, or Se and $R_1 \ R_2$ and R_3 are as above

The different groups L may be the same or different quinolate derivatives such as

where R is hydrocarbyl, aliphatic, aromatic or heterocyclic carboxy, aryloxy, hydroxy or alkoxy e.g. the 8 hydroxy quinolate derivatives or

$$R \xrightarrow{O^{-}} B \xrightarrow{O^{-}} O \xrightarrow{R_{1}} P \xrightarrow{O^{-}} O \xrightarrow{(IX)} O \xrightarrow{(IX)} O \xrightarrow{(X)} O \xrightarrow{R_{2}} P \xrightarrow{O^{-}} O \xrightarrow{R_{2}} O$$

where R, R_1 , and R_2 are as above or are H or F e.g. R_1 and R_2 are alkyl or alkoxy groups

As stated above the different groups L may also be the same or different carboxylate groups e.g.

$$R_5$$
— C
(XIII)

where R_5 is a substituted or unsubstituted aromatic, polycyclic or heterocyclic ring a polypyridyl group, R_5 can also be a 2-ethyl hexyl group so L_n is 2-ethylhexanoate or R_5 can be a chair structure so that L_n is 2-acetyl cyclohexanoate or $L\alpha$ can be

where R is as above e.g. alkyl, allenyl, amino or a fused ring such as a cyclic or polycyclic ring.

5 The different groups L may also be

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Where R, R_1 and R_2 are as above.

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The invention also provides an electroluminescent device which comprises (i) a first electrode, (ii) a layer of a electroluminescent compound as described above and (iii) a second electrode.

The first electrode can function as the cathode and the second electrode can function as the anode and preferably there is a layer of a hole transporting material between the anode and the layer of the electroluminescent compound.

The hole transporting material can be any of the hole transporting materials used in electroluminescent devices.

The hole transporting material can be an amine complex such as poly (vinylcarbazole), N, N'-diphenyl-N, N'-bis (3-methylphenyl) -1,1' -biphenyl -4,4'-diamine (TPD), an unsubstituted or substituted polymer of an amino substituted aromatic compound, a polyaniline, substituted polyanilines, polythiophenes, substituted polythiophenes, polysilanes etc. Examples of polyanilines are polymers of

where R is in the ortho – or meta-position and is hydrogen, C1-18 alkyl, C1-6 alkoxy, amino, chloro, bromo, hydroxy or the group

where R is alky or aryl and R' is hydrogen, C1-6 alkyl or aryl with at least one other monomer of formula I above.

Or the hole transporting material can be a polyaniline, polyanilines which can be used in the present invention have the general formula

$$\begin{array}{c|c}
 & R \\
 & N \\$$

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where p is from 1 to 10 and n is from 1 to 20, R is as defined above and X is an anion, preferably selected from Cl, Br, SO₄, BF₄, PF₆, H₂PO₃, H₂PO₄, arylsulphonate, arenedicarboxylate, polystyrenesulphonate, polyacrylate alkysulphonate, vinylsulphonate, vinylbenzene sulphonate, cellulose sulphonate, camphor sulphonates, cellulose sulphate or a perfluorinated polyanion.

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Examples of arylsulphonates are p-toluenesulphonate, benzenesulphonate, 9,10-anthraquinone-sulphonate and anthracenesulphonate, an example of an arenedicarboxylate is phthalate and an example of arenecarboxylate is benzoate.

We have found that protonated polymers of the unsubstituted or substituted polymer of an amino substituted aromatic compound such as a polyaniline are difficult to evaporate or cannot be evaporated, however we have surprisingly found that if the unsubstituted or substituted polymer of an amino substituted aromatic compound is deprotonated the it can be easily evaporated i.e. the polymer is evaporable.

Preferably evaporable deprotonated polymers of unsubstituted or substituted polymer of an amino substituted aromatic compound are used. The de-protonated unsubstituted or substituted polymer of an amino substituted aromatic compound can be formed by deprotonating the polymer by treatment with an alkali such as ammonium hydroxide or an alkali metal hydroxide such as sodium hydroxide or potassium hydroxide.

The degree of protonation can be controlled by forming a protonated polyaniline and de-protonating. Methods of preparing polyanilines are described in the article by A. G. MacDiarmid and A. F. Epstein, Faraday Discussions, Chem Soc.88 P319 1989.

The conductivity of the polyaniline is dependant on the degree of protonation with the maximum conductivity being when the degree of protonation is between 40 and 60% e.g. about 50% for example.

Preferably the polymer is substantially fully deprotonated

A polyaniline can be formed of octamer units i.e. p is four e.g.

The polyanilines can have conductivities of the order of 1×10^{-1} Siemen cm⁻¹ or higher.

The aromatic rings can be unsubstituted or substituted e.g. by a C1 to 20 alkyl group such as ethyl.

The polyaniline can be a copolymer of aniline and preferred copolymers are the copolymers of aniline with o-anisidine, m-sulphanilic acid or o-aminophenol, or o-toluidine with o-aminophenol, o-ethylaniline, o-phenylene diamine or with amino anthracenes.

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Other polymers of an amino substituted aromatic compound which can be used include substituted or unsubstituted polyaminonapthalenes, polyaminoanthracenes, polyaminophenanthrenes, etc. and polymers of any other condensed polyaromatic compound. Polyaminoanthracenes and methods of making them are disclosed in US Patent 6,153,726. The aromatic rings can be unsubstituted or substituted e.g. by a group R as defined above.

Other hole transporting materials are conjugated polymer and the conjugated polymers which can be used can be any of the conjugated polymers disclosed or referred to in US 5807627, PCT/WO90/13148 and PCT/WO92/03490.

The preferred conjugated polymers are poly (p-phenylenevinylene)-PPV and copolymers including PPV. Other preferred polymers are poly(2,5 dialkoxyphenylene vinylene) such as poly (2-methoxy-5-(2-methoxypentyloxy-1,4-phenylene vinylene), poly(2-methoxypentyloxy)-1,4-phenylenevinylene), poly(2-methoxy-5-(2-dodecyloxy-1,4-phenylenevinylene) and other poly(2,5 dialkoxyphenylenevinylenes) with at least one of the alkoxy groups being a long chain solubilising alkoxy group,

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poly fluorenes and oligofluorenes, polyphenylenes and oligophenylenes, polyanthracenes and oligo anthracenes, ploythiophenes and oligothiophenes.

In PPV the phenylene ring may optionally carry one or more substituents e.g. each independently selected from alkyl, preferably methyl, alkoxy, preferably methoxy or ethoxy.

Any poly(arylenevinylene) including substituted derivatives thereof can be used and the phenylene ring in poly(p-phenylenevinylene) may be replaced by a fused ring system such as anthracene or naphthlyene ring and the number of vinylene groups in each polyphenylenevinylene moiety can be increased e.g. up to 7 or higher.

The conjugated polymers can be made by the methods disclosed in US 5807627, PCT/WO90/13148 and PCT/WO92/03490.

The thickness of the hole transporting layer is preferably 20nm to 200nm.

The polymers of an amino substituted aromatic compound such as polyanilines referred to above can also be used as buffer layers with or in conjunction with other hole transporting materials.

The structural formulae of some other hole transporting materials are shown in Figures 4, 5, 6, 7 and 8 of the drawings, where R₁, R₂ and R₃ can be the same or different and are selected from hydrogen, and substituted and unsubstituted hydrocarbyl groups such as substituted and unsubstituted aliphatic groups, substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorocarbons such as trifluoryl methyl groups, halogens such as fluorine or thiophenyl groups; R₁, R₂ and R₃ can also form substituted and unsubstituted fused aromatic, heterocyclic and polycyclic ring structures and can be copolymerisable with a monomer e.g. styrene. X is Se, S or O, Y can be hydrogen, substituted or unsubstituted hydrocarbyl

m Complexes 2 11:39

groups, such as substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorine, fluorocarbons such as trifluoryl methyl groups, halogens such as fluorine or thiophenyl groups or nitrile.

- Examples of R₁ and/or R₂ and/or R₃ include aliphatic, aromatic and heterocyclic alkoxy, aryloxy and carboxy groups, substituted and substituted phenyl, fluorophenyl, biphenyl, phenanthrene, anthracene, naphthyl and fluorene groups alkyl groups such as t-butyl, heterocyclic groups such as carbazole.
- Optionally there is a layer of an electron injecting material between the anode and the electroluminescent material layer, the electron injecting material is a material which will transport electrons when an electric current is passed through electron injecting materials include a metal complex such as a metal quinolate e.g. an aluminium quinolate, lithium quinolate, a cyano anthracene such as 9,10 dicyano anthracene, cyano substituted aromatic compounds, tetracyanoquinidodimethane a polystyrene sulphonate or a compound with the structural formulae shown in figures 2 or 3 of the drawings in which the phenyl rings can be substituted with substituents R as defined above.
- The cathode is preferably a transparent substrate such as a conductive glass or plastic material which acts as the anode, preferred substrates are conductive glasses such as indium tin oxide coated glass, but any glass which is conductive or has a conductive layer such as a metal or conductive polymer can be used. Conductive polymers and conductive polymer coated glass or plastics materials can also be used as the substrate.

The anode is preferably a low work function metal e.g. aluminium, calcium, lithium, silver/magnesium alloys, rare earth metal alloys etc., aluminium is a preferred metal.

A metal fluoride such as an alkali metal, rare earth metal or their alloys can be used as the second electrode for example by having a metal fluoride layer formed on a

- 16 -

metal.

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Claims

- 1. An electroluminescent compound of formula $M(L)_n$ and $MO(L)_{n-2}$ where M is a metal in a valency state n of greater than 3 and L is an organic ligand, the ligands L can be the same or different e.g. $M(L_1)$ (L_2) (L_3) (L_4)... or $MO(L_1)$ (L_2)....
- 2. An electroluminescent compound as claimed in claim 1 in which the metal M is a titanium, zirconium or hafnium in the four valency state or vanadium, niobium or tantulum in the five valency state.
- 3. An electroluminescent compound as claimed in claim 1 of formula of fig. 1.
- 4. An electroluminescent compound as claimed in any one of claims 1 to 3 in which the ligands L are the same or different.
- 5. An electroluminescent compound as claimed in claim 1 of formula M(L₁)(L₂)(L₃)(L₄) or MO(L₁) (L₂) where the groups (L₁), (L₂), (L₃) and (L₄) are the same or different where M is zirconium, hafnium or titanium in the four valency state.
 - 6. An electroluminescent compound as claimed in claim 1 of formula $M(L_1)(L_2)(L_3)(L_4)(L_5)$ or $MO(L_1)$ (L_2) (L_3) where the groups (L_1), (L_2), (L_3) and (L_4) (L_5) are the same or different where M is vanadium, niobium or tantulum in the five valency state.
 - 7. An electroluminescent compound as claimed in any one of the preceding claims in which the ligands L are selected from ligands (I) to (XVIII) herein.
- 8. An electroluminescent compound as claimed in any one of the preceding claims in which the ligands L are selected from quinolate, porphoryin, pyrazalones, and β diketones such as dibenzoyl methane.

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- 9. An electroluminescent composition which comprises an electroluminescent compound as claimed in claim 1 or claim 2 and a dopant.
- 10. An electroluminescent composition as claimed in claim 9 in which the dopant is selected from diphenylacridine, coumarins, perylene, quinolates, porphoryin, porphines, pyrazalones and their derivatives.
 - 11. An electroluminescent composition as claimed in claim 9 in which the dopant is selected from compounds of formula (A), (B) and (C) and compounds of figures 9 to 13 of the drawings.
 - 12. An electroluminescent device which comprises (i) a first electrode, (ii) a layer of electroluminescent compound or composition as claimed in any one of claims 1 to 11 and (iii) a second electrode.
 - 13. An electroluminescent device as claimed in claim 12 in which there is a layer of a hole transmitting material between the first electrode and the electroluminescent layer.
- 20 14. An electroluminescent device as claimed in claim 13 in which the hole transmitting material is an aromatic amine complex.
 - 15. An electroluminescent device as claimed in claim 12 in which the hole transmitting material is polyaromatic amine complex.
 - 16. An electroluminescent device as claimed in claim 13 in which the hole transmitting material is a film of a polymer selected from poly(vinylcarbazole), N,N'-diphenyl-N,N'-bis (3-methylphenyl) -1,1' -biphenyl -4,4'-diamine (TPD), polyaniline, substituted polyanilines, polythiophenes, substituted polythiophenes, polysilanes and substituted polysilanes.

m Complexes 2 11:39

- 17. An electroluminescent device as claimed in claim 13 in which the hole transmitting material is a film of a compound of formula (XIX) or (XXV) herein or as in figures 4 to 8 of the drawings.
- 18. An electroluminescent device as claimed in claim 14 in which the hole transmitting material is a copolymer of aniline, a copolymer of aniline with o-anisidine, m-sulphanilic acid or o-aminophenol, or o-toluidine with o-aminophenol, o-ethylaniline, o-phenylene diamine or with an amino anthracene.
- 19. An electroluminescent device as claimed in claim 15 in which the hole transmitting material is a conjugated polymer.
- 20. An electroluminescent device as claimed in claim 18 in which the conjugated polymer is selected from poly (p-phenylenevinylene)-PPV and copolymers including 15 PPV, poly(2,5)dialkoxyphenylene vinylene), poly (2-methoxy-5-(2methoxypentyloxy-1,4-phenylene vinylene), poly(2-methoxypentyloxy)-1,4phenylenevinylene), poly(2-methoxy-5-(2-dodecyloxy-1,4-phenylenevinylene) and other poly(2,5 dialkoxyphenylenevinylenes) with at least one of the alkoxy groups being a long chain solubilising alkoxy group, poly fluorenes and oligofluorenes, polyphenylenes and oligophenylenes, polyanthracenes and oligo anthracenes, 20 ploythiophenes and oligothiophenes.
 - 21. An electroluminescent device as claimed in any one of claims 12 to 20 in which the electroluminescent compound is mixed with the hole transmitting material.
 - 22. An electroluminescent device as claimed in any one of claims 14 to 20 in which there is a layer of an electron transmitting material between the cathode and the electroluminescent compound layer.

- 23. An electroluminescent device as claimed in claim 22 in which the electron transmitting material is a metal quinolate.
- 24. An electroluminescent device as claimed in claim 23 in which the metal quinolate is an aluminium quinolate or lithium quinolate
 - 25. An electroluminescent device as claimed in claim 24 in which the electron transmitting material is of formula Mx(DBM)_n where Mx is a metal and DBM is dibenzoyl methane and n is the valency of Mx.
 - 26. An electroluminescent device as claimed in claim 22 in which the electron transmitting material is a cyano anthracene such as 9,10 dicyano anthracene, a polystyrene sulphonate or a compound of formulae shown in figure 2 or 3 of the drawings.
 - 27. An electroluminescent device as claimed in any one of claims 22 to 24 in which the electron transmitting material is mixed with the electroluminescent compound.
- 28. An electroluminescent device as claimed in any one of the claims 12 to 25 in which the first electrode is a transparent electricity conducting glass electrode.
 - 29. An electroluminescent device as claimed in any one of the claims 12 to 26 in which the second electrode is selected from aluminium, calcium, lithium, magnesium and alloys thereof and silver/magnesium alloys.

Abstract

An electroluminescent compound is a organometallic complex in which the metal is titanium, zirconium, hafnium, vanadium, niobium or tantulum.

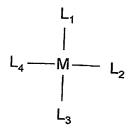


Fig. 1a

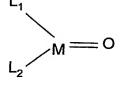
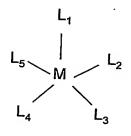


Fig. 1b



. Fig. 1c

Fig. 1d

2/13

Alq

Bebq

BAlq1 ·

ZnPBO

ZnPBT

DTVb1

Fig. 2

Fig. 3

OXD- Star

Fig. 4

Fig. 5

6/13

$$R_1$$
 R_2 R_3 R_4

Fig. 14a

Fig. 14b

$$R_1$$
 R_2
 S
 S
 R_3
 R_4
or

$$R_1$$
 R_2
 S
 S
 S
 S
 S
 R_3
 R_4

Fig. 6

Fig. 7

8/13

mTADATA

Fig. 8

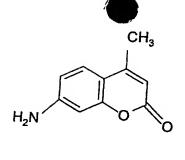


Fig. 17

$$R_1$$
 R_2 R_3 R_3 R_4 R_4 R_5 R_5 R_6 R_6

$$R_1$$
 R_2 R_3

Fig. 19

Fig. 21

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